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LLNL-PROC-648838

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January 27, 2014

LTD 15
Pasadena, CA, United States
June 24, 2013 through June 28, 2013

Disclaimer

Journal of Low Temperature Physics manuscript No.
(will be inserted by the editor)

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Received: date / Accepted: date

Abstract Non-destructive assay (NDA) of nuclear materials would benefit from gamma detectors with improved energy resolution in cases where line overlap in current Ge detectors limits NDA accuracy. We are developing metallic magnetic calorimeter (MMC) gamma-detectors for this purpose by electroplating $\sim$150 μm thick Au absorbers into microfabricated molds on top of Au:Er sensors. Initial tests under non-optimized conditions show an energy resolution of $\sim$200 eV FWHM at 60 keV. Monte Carlo simulations illustrate that this resolution is starting to be sufficient for direct detection of ^{242}Pu in plutonium separated from spent nuclear fuel.

Keywords MMC · gamma ray

1 Introduction

One of the primary tools used in nuclear safeguards to verify the operating history of a nuclear reactor is to compare the operator declarations to the measured spent fuel characteristics. This is typically done using ratios of high intensity gamma ray lines with a high-purity germanium (HPGe) detector. This method is becoming less reliable as advances in materials engineering enable higher burn-up of the nuclear fuel before failure. As the fuel assembly is irradiated for longer periods of time, the correlation of the high intensity gamma ray lines to fuel characteristics weakens. In addition, high burn-up produces increasing amounts of ^{242}Pu, which can currently only be measured indirectly [1]. If the ratios of all Pu isotopes could

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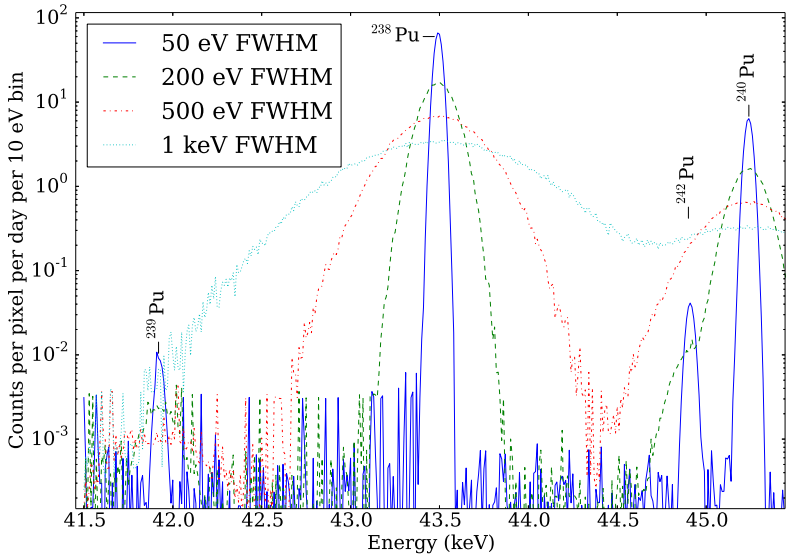

Fig. 1 Geant4 Simulation of Pu gamma ray lines near 40 keV for different detector resolutions, assuming a Pu point source and full separation of Pu from the fission and activation products of the spent fuel. (color figure online)

be determined more accurately, better constraints could be placed on initial fuel enrichment and reactor operating history. This makes the development of high-resolution gamma detectors desirable for use in this application.

Cryogenic gamma detectors with operating temperatures of $\sim$0.1 K have been developed for several decades. Different detector technologies have been used to attain an energy resolution of $<$100 eV FWHM, including Si and Ge thermistors[2,3], superconducting transition edge sensors (TESs)[4,5], and more recently, metallic magnetic calorimeters[6]. We are focusing on MMC gamma detectors, because they offer higher energy resolution and potentially higher count rate capabilities than other technologies. Specifically, MMC signals decay with a single thermal time constant[7]. This should make them compatible with modern digital pulse processing algorithms to read them out under pile-up conditions at several hundred counts/s with minimal loss in energy resolution[8]. Here we discuss our recent developments of MMC gamma detectors for nuclear safeguards applications.

2 Direct measurement of ^{242}Pu in spent fuel

In order to measure the different Pu isotopes it is currently necessary to chemically separate the Pu from other metals. This is already common for mass spectrometry and it is also a standard part of fuel reprocessing. The separated Pu is then typically deposited onto a thin Al planchet to minimize the scattering in the source and placed in front of gamma detector for counting.

We have performed Geant4 Monte Carlo simulations to determine the resolution requirements of our detector[9]. As an input source we used the program

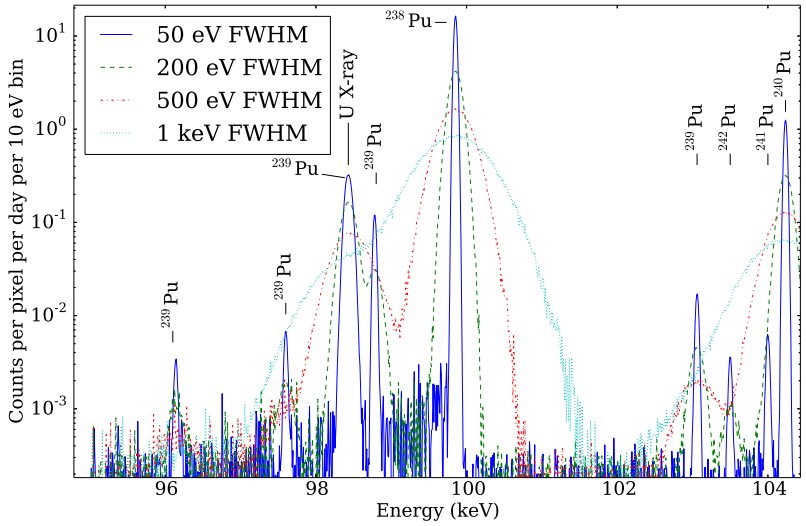

Fig. 2 Geant4 Simulation of Pu gamma ray lines near 100 keV for different detector resolutions, assuming a Pu point source and full separation of Pu from the fission and activation products of the spent fuel. (color figure online)

ORIGEN-ARP to generate the expected Pu content for a fuel assembly with a typical irradiation profile for a modern pressurized water reactor. In order to simplify the simulation we assumed a point source geometry and perfect Pu seperation from other elements. We focused on the regions around 40 keV and 100 keV, since they contain gamma rays from all relevant Pu isotopes, and the proximity of the lines enables ratio measurements with a simple correction for efficiency. In order to calculate the counts per pixel per day we assumed that the source strength was such that the detector could be operated at 100 counts/s.

Figure 1 shows the simulated spectrum in the 40 keV region with several different detector resolutions. The 1 keV FWHM spectrum is typical of a commercial coaxial HPGe detector, while the 500 eV FWHM spectrum is typical of a thin planar HPGe optimized for low energy spectroscopy. The 200 eV FWHM spectrum shows the limit at which the ^{242}Pu line at 44.8 keV can be resolved, which is comparable to the best MMC spectrum in this work. Finally, the 50 eV FWHM is comparable to what is theoretically achievable in an optimally designed MMC that would have appreciable efficiency at 40 keV. Based on statistics, we estimate that a resolution of $\sim$75 eV FWHM is required to determine the Pu-242 content to 1% precision, because the interfering Pu-240 line is roughly 100 times stronger and separated by only 300 eV[10]. This precision depends on the Pu-242 / Pu-240 ratio and the Compton background level, which may be higher than in Figure 1 because of the simplifying assumptions made in the simulations.

Figure 2 shows the simulated spectra for the same four detectors in the 100 keV region, which is widely used for Pu analysis[11]. Again, an MMC energy resolution of 200 eV starts to separate the individual Pu emissions, although it may be more difficult to measure ^{242}Pu in this region even with 50 eV FWHM. This

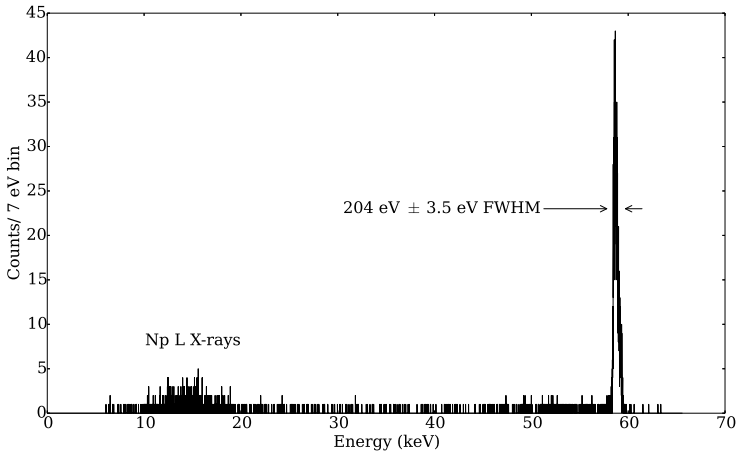

Fig. 3 Spectra of ^{241}Am taken with a maXs-200 MMC.

is because the line is an order of magnitude weaker, and the secondary Pu $K_{\alpha 1}$ X-ray fluorescence at 103.75 keV can provide strong interference in concentrated Pu samples.

3 Gamma-ray measurements of ^{241}Am

In order to demonstrate the feasibility of the Pu measurement with an MMC we installed an ^{241}Am source in front of a Heidelberg maXs-200 MMC in a 18 mK dilution fridge operated at the Kirchhoff Institute for Physics[6]. This design uses a Au:Er sensor deposited on top of a 0.5 mm $\times$ 2 mm meander of Nb, which is used to both apply the field and readout the signal. In order to be sensitive to hard X-rays a 150 μm thick Au absorber is electroplated on top of the Au:Er. This device was read out using a two-stage SQUID, model C6XS116 from Magnicon GmbH. The raw pulses were saved using a 12-bit Gage data acquisition card, and processed using an optimal filter routine. Despite wiring issues with the device that forced us to operate with approximately 10% of the designed magnetizing current it was still possible to achieve a resolution of 200 eV at 60 keV (Figure 3). This performance shows that it should be possible to directly measure the ^{242}Pu line using this device even in sub-optimal operating conditions. Under optimal conditions devices similar to this one have achieved an energy resolution of 60 eV FWHM at 60 keV[6]. We are currently working on attaining similar resolution.

4 Outlook

High accuracy isotopic analysis of plutonium separated from spent nuclear fuel will eventually require the development of MMC arrays to collect spectra with sufficiently high statistics. While the required array size will depend on the specific sample and the accuracy requirements, we can make a rough estimate for Pu

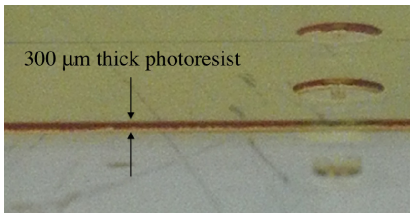

Fig. 4 300 μm thick AZ-125 nXT photoresist on test slide. (color figure online)

from spent fuel. In high burn-up samples with a ^{242}Pu concentration of $\sim$10%, the signal from this isotope ($t_{1/2}$ = 373300 y) will constitute only $\sim$0.1% of the total gamma flux [11], which will be dominated by the shorter-lived isotopes of ^{238}Pu ($t_{1/2}$ = 88 y) and ^{241}Pu ($t_{1/2}$ = 14 y). An accuracy of order $\sim$1%, which requires a total of 10^4 counts in the ^{242}Pu line, thus requires a spectrum with a total of $\sim 10^7$ counts. Since the MMC signal decay time is $\sim$1 ms and the count rate per pixel is therefore limited to at most a few 100 counts/s, a single MMC detector would have to count $>10^5$ s, or several days. However, a 100-pixel MMC array would reduce this time to a few hours, which is acceptable for safeguards applications.

Our fabrication of MMC arrays is currently limited by the difficulty in removing the SU-8 mold for the Au absorbers, which tends to stick to the MMCs and limit the device yield. We are therefore developing a mold based on AZ-125 nXT photoresist, which can be patterned with very high aspect ratios [12]. We have successfully spin coated AZ-125 nXT to a thickness of 300 μm, and have shown that we can produce thick molds for electroplating without the removal problems associated with SU-8 (Figure 4). This should simplify the fabrication of high-yield MMC arrays with thick electroplated absorbers in the future.

Acknowledgements This work was funded by the U.S. Department of Energy Office on Non-Proliferation Research NA-22 under grant LL12-MagMicro-PD2Ja. This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344.

References

1. G. Bignan, W. Ruhter, H. Ottmar, A. Schubert, and C. Zimmerman. Plutonium Isotopic Determination By Gamma Spectrometry: Recommendations for the ^{242}Pu content evaluation using a new algorithm. *ESARDA BULLETIN* **28**:16, (1998)
2. S.H. Moseley, R.L. Kelley, R.J. Schoelkopf and A.E. Szymkowiak. Advances Toward High Spectral Resolution Quantum X-Ray Calorimetry. *IEEE TNS* **35**:1, 59-64 (1988)
3. A. Alessandrello, J.W. Beeman, C. Brofferio, O. Cremonesi, E. Fiorini, A. Giuliani, E.E. Haller, A. Monfardini, A. Nucciotti, M. Pavan, G. Pessina, E. Previtali and L. Zanotti. High Energy Resolution Bolometers for Nuclear Physics and X-Ray Spectroscopy. *Physical Review Letters* **82**:3, 513-515 (1999)

4. M. F. Cunningham, J.N. Ullom, T. Miyazaki, S.E. Labov, J. Clarke, T.M. Lanting, A.T. Lee, P.L. Richards, J. Yoon and H. Spieler. High-resolution operation of frequency-multiplexed transition-edge photon sensors. *Applied Physics Letters* **81**:1, 159-161 (2002)
5. B.L. Zink, J.N. Ullom, J.A. Beall, K.D. Irwin, W.B. Doriese, W.D. Duncan, L. Ferreira, G.C. Hilton, R.D. Horansky, C.D. Reintsema and L.R. Vale. Array-compatible transition-edge sensor microcalorimeter γ-ray detection with 42 eV energy resolution at 103 keV. *Applied Physics Letters* **89**, 124101 (2006)
6. C. Pies, S. Schaefer, S. Heuser, S. Kempf, A. Pabinger, J.-P. Porst, P. Ranitsch, N. Foerster, D. Hengstler, A. Kampkoetter, T. Wolf, L. Gastaldo, A. Fleischmann, and C. Enss. maXs: Microcalorimeter Arrays for High-Resolution X-Ray Spectroscopy at GSI/FAIR, *J. Low Temp. Phys.* **167**, 269 (2012)
7. A. Fleishmann, L. Gastaldo, S. Kempf, A. Kirsch, A. Pabinger, C. Pies, J.-P. Porst, P. Ranitzsch, S. Schäfer, F.v. Seggern, T. Wolf, C. Enss, and G.M. Seidel. Metallic magnetic calorimeters, *AIP Conf. Proc.* **1185**, 571-578 (2009)
8. P. A.B. Scoullar, C.C. McLean, and R.J. Evans. Real Time Pulse Pileup Recovery in a High Throughput Digital Pulse Processor, *AIP Conf. Proc.* **1412**, 271-277 (2011)
9. S. Agnostinelli et al., Geant4: a simulation toolkit, *NIM A* **503**:3, 250-303 (2003)
10. O.B. Drury, S.F. Terracol, and S. Friedrich, Quantifying the benefits of ultra-high energy resolution for gamma ray spectrometry, *Phys. Stat. Sol. C* **2**, 1468-1479 (2005)
11. D. Reilly, N. Ensslin, H. Smith, Jr., and S. Kreiner, *Passive Nondestructive Assay of Nuclear Materials* , 221-267. National Technical Information Service, Springfield, VA (1991)
12. M. Staab, F. Greiner, M. Schlosser, and H.F. Schlaak, Applications of Novel High-Aspect-Ratio Ultrathick UV Photoresist for Microelectroplating, *JMEMS Letters* **20**:4, 794-796 (2011)